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Synchrotron X-ray diffuse scattering studies of PMN-PT relaxor ferroelectrics

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Introduction: Dielectric properties of $[Pb(Mg_{1/3}Nb_{2/3})O_3]_{1-x}$-$[PbTiO_3]_x$ (PMN-xPT) solid solutions strongly depend on the amount of $PbTiO_3$ (PT) doping. For example, compositions with x<0.32 exhibit strong frequency dispersion of dielectric susceptibility common for relaxor ferroelectrics. However, no such frequency dispersion, typical for "normal ferroelectrics", is observed in compositions with x>0.32. Dielectric relaxation in PMN-PT is commonly explained by the competition between randomly occurring dynamic ferroelectric and anti-ferroelectric fluctuations in the presence of the underlying matrix disorder [1]. Anti-ferroelectric fluctuations are formed by locally correlated atomic displacements that change linear dimensions of the unit cell in certain crystallographic directions. These displacements produce superlattice reflections that were studied with single crystal x-ray diffuse scattering techniques.

Methods and Materials: Single crystals of PMN-xPT in the shape of rectangular platelets with linear dimensions 5x5x1 mm^3 were investigated in the closed cycle He gas DISPLEX cryostat mounted on the standard four-circle HUBER diffractometer. Distribution of the diffracted intensity in the reciprocal space has been mapped using computer automated 2D hkl scans in the reciprocal planes where superlattice reflections were expected to exist. Energy of the incident synchrotron radiation was tuned to 10 keV by two Si (111) crystals with subsequent focusing by Si cylindrical mirror. Diffracted intensity was measured with a KEVEX Si solid-state detector. This experimental setup completely eliminated data contamination by second and higher order x-ray harmonics. Temperature dependence of the superlattice reflections was followed from 10 K up to room temperature.

Results: Fig. 1 shows distribution of the diffuse scattering intensity in the reciprocal space near (022) Bragg peak for several PMN-xPT compositions. Reflections marked F and α on the plot are superlattice peaks at different Brillouine zone boundaries. Detailed analysis of the temperature dependence has shown that F peaks are primarily due to Nb/Mg chemical ordering, while α superlattice peaks have pure atomic displacement origin [2, 3]. Integrated intensity of the α peaks exhibited strong temperature dependence near phenomenological Vogel-Fulcher freezing temperature obtained from macroscopic dielectric relaxation measurements for different chemical compositions [2, 3]. It was found that no superlattice reflections exist in PMN-0.32PT, which is "normal ferroelectric" crystal. Studies of the α peaks provide us with better understanding of the structure-property relationship in PMN-PT. The α superlattice reflections are believed to originate from locally correlated (~30 Å) oxygen octahedral rotations and Pb displacements. It is possible that fluctuations created by these displacements disturb long-range polar ferroelectric ordering and lead to glassy relaxor behavior.

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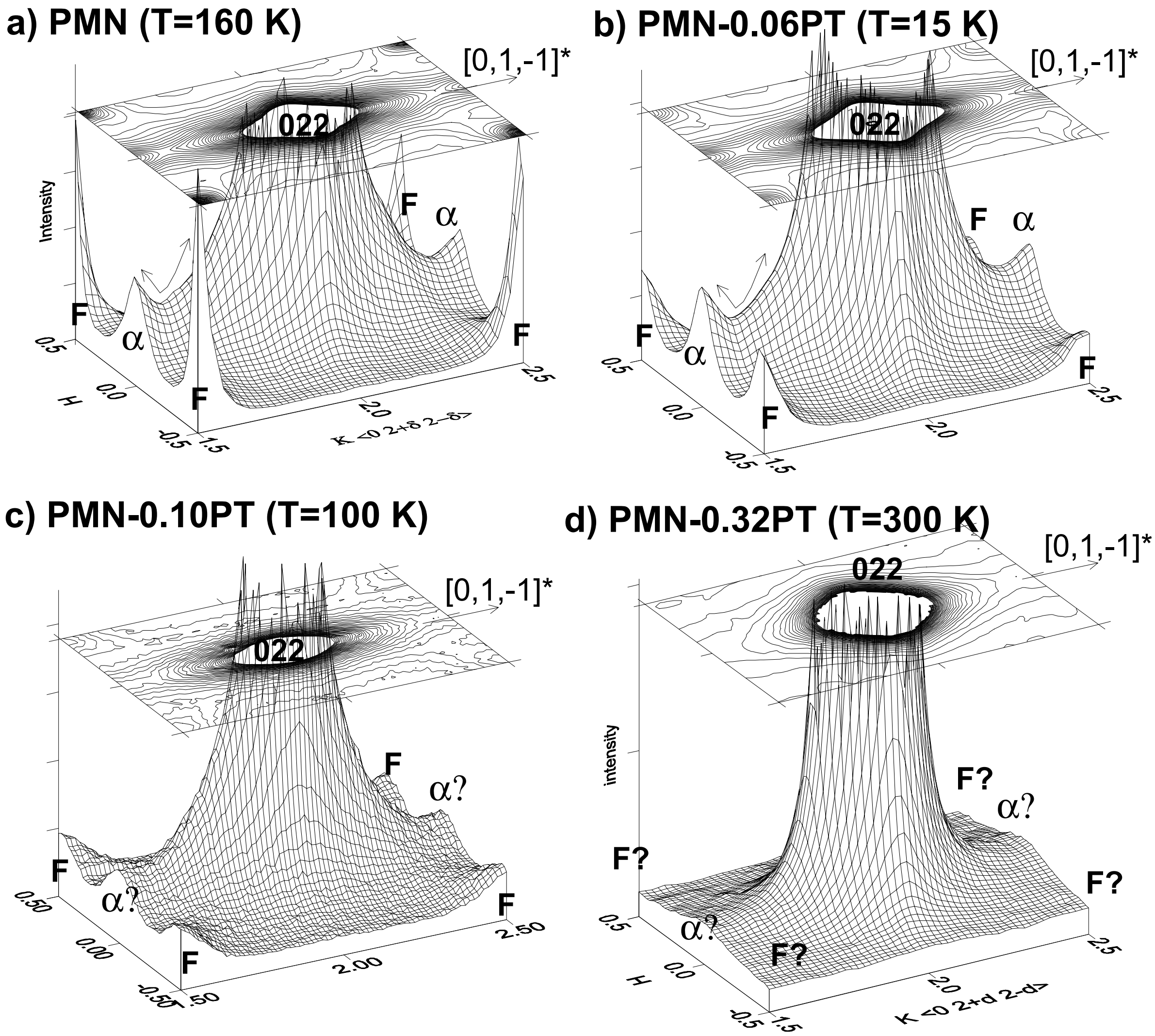

Figure1 : Distribution of diffuse scattering intensity near (022) Bragg peak and superlattice reflections marked α and F in PMN-xPT for x=0, 0.06, 0.1, 0.32.